

Argonne National Laboratory

**SOME APPLICATIONS OF
SCINTILLATION SPECTROMETRY
TO NONDESTRUCTIVE TESTING**

by

Ronald B. Perry

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I. INTRODUCTION

Gamma-ray scintillation spectrometry provides a nondestructive method of reactor fuel assay for U^{235} content which is useful for quality

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ABSTRACT

Gamma-ray scintillation spectrometry is a useful tool for the nondestructive assay of the U^{235} content of research reactor fuel. The U^{235} is the only isotope of uranium that produces a strong peak of gamma energy at 184 keV. Accordingly, the U^{235} content of reactor fuel can be accurately determined by counting the 184-keV gamma emission.

Uranium foil standards of known weight and isotopic content can be used for assay of fabricated reactor fuel with a product of density and thickness small enough to permit accurate measurement of the self-absorption factor. Two techniques for nondestructive assay of fabricated fuel in flat configurations, based on the use of uranium foils as standards, are discussed herein. In the first method a small area is counted and homogeneity is assumed. In the second method, a two-dimensional scanning technique is employed such that the entire sample is counted. A similar scanning technique has been used for tubes of 5.08- to 7.62-cm diameter.

Other configurations of reactor fuel have products of density and thickness or geometrical shapes that do not permit accurate measurement of the self-absorption factor for the 184-keV gamma. Standard samples for these configurations may be taken from the material itself and standardized by mass-spectrometric and chemical analysis or specially fabricated from material of known U^{235} content.

Scintillation counting techniques may be applied to other reactor fuel testing problems. Distribution of U^{235} in dispersion and alloy-type fuels can be measured by counting small areas with a well-collimated detector.

I. INTRODUCTION

Gamma-ray scintillation spectrometry provides a nondestructive method of reactor fuel assay for U^{235} content which is useful for quality

control and accountability purposes. In a sample of highly enriched uranium containing 93% U^{235} , 5.5% U^{238} , 1% U^{234} , and 0.5% U^{236} , the U^{235} isotope contributes only 3% of the total disintegrations. But examination of the gamma-ray spectra of various uranium isotopes (see Figures 1a, 1b, 1c, and 1d) indicates that the U^{235} isotope is the primary source of gamma radiation of 184-keV energy. In normal uranium the U^{235} isotope produces approximately 2% of the disintegrations, but its presence is still detectable from the gamma-ray spectrum (see Figure 1b). The intense peak of gamma energy at 184 keV from the decay of U^{235} is especially useful for U^{235} assay. Procedures for quantitative measurement of U^{235} content in reactor fuels based on counting the 184-keV gamma from U^{235} by means of a single-channel scintillation spectrometer with a NaI(Tl) crystal detector have been established.

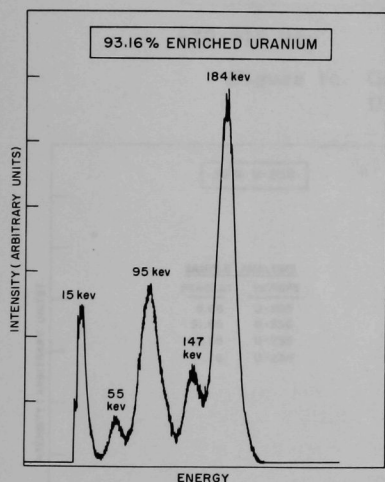


Figure 1a

Gamma-ray Spectrum of
Uranium Enriched in U^{235}

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GAMMA SPECTRUM - NORMAL URANIUM

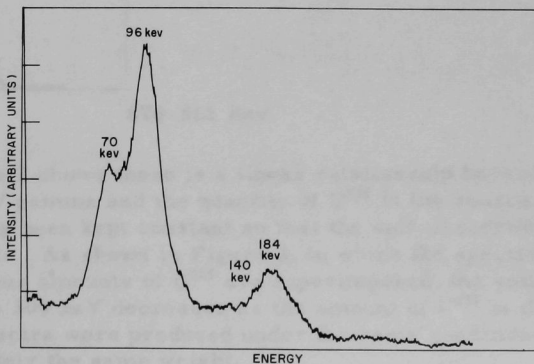
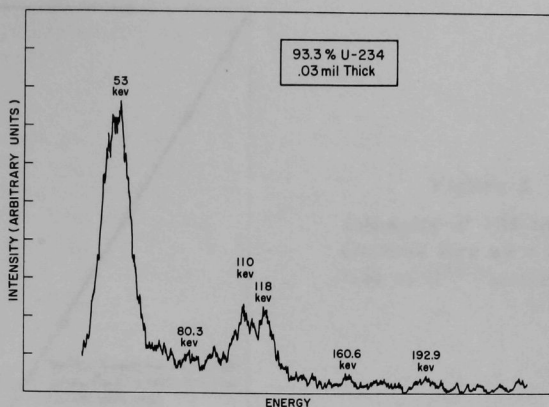


Figure 1b

Gamma Spectrum -
Normal Uranium

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Figure 1c. Gamma-ray Spectrum of
Uranium Enriched in U^{234}

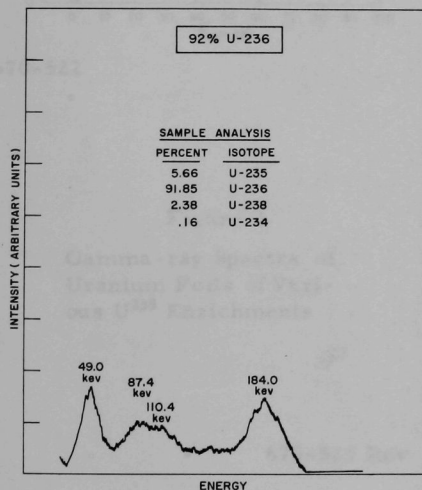


Figure 1d

Gamma-ray Spectrum of
Uranium Enriched in U^{236}

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The graph in Figure 2 shows there is a linear relationship between the intensity of the 184-keV gamma and the quantity of U^{235} in the source. The quantity of uranium has been kept constant so that the self-absorption is the same for each sample. As shown in Figure 3, in which the spectra of sources containing various amounts of U^{235} are superimposed, the entire gamma-ray spectrum up to 300 keV decreases as the amount of U^{235} in the source decreases. The spectra were produced under the same conditions with sources of approximately the same weight.

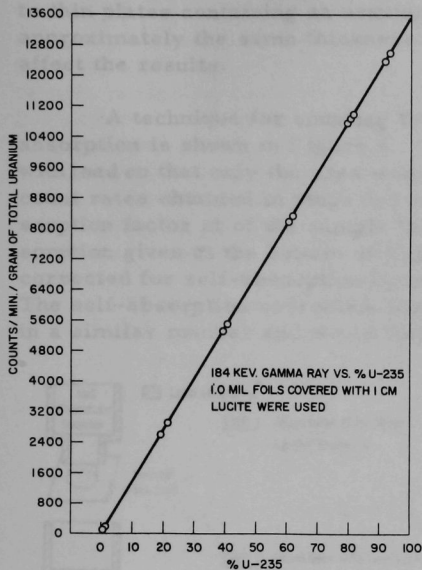


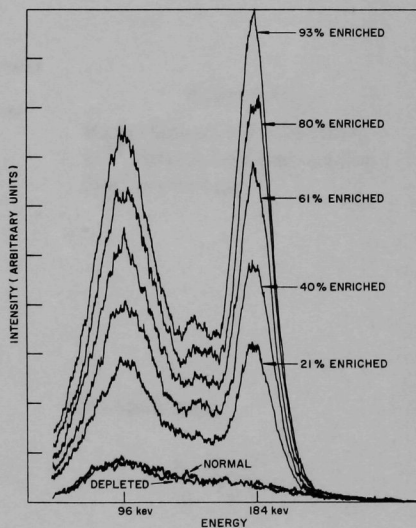
Figure 2

Intensity of 184-keV
Gamma Ray as a Func-
tion of U^{235} Enrichment

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Figure 3

Gamma-ray Spectra of
Uranium Foils of Vari-
ous U^{235} Enrichments



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II. U^{235} ASSAY PROCEDURE

A. Static Method

Fuel configurations with density-times-thickness products small enough to permit accurate measurement of the self-absorption factor for the 184-keV γ radiation from U^{235} can be compared with standard uranium foils of known weight and isotopic content.⁽¹⁾ This technique is applicable

to thin plates containing an uranium alloy or dispersion. Cladding of approximately the same thickness on both sides of the sample will not affect the results.

A technique for counting the samples and measuring the self-absorption is shown in Figure 4. The detector is shielded and collimated with lead so that only the area under the collimator is counted. The three count rates obtained in steps 1-3 in Figure 4 are used to calculate the absorption factor μt of the sample from the equation for exponential absorption given at the bottom of Figure 4. The count rate of the sample is corrected for self-absorption by means of the equation given in Figure 5. The self-absorption correction for the standard foil would be determined in a similar manner and would then be a known constant.

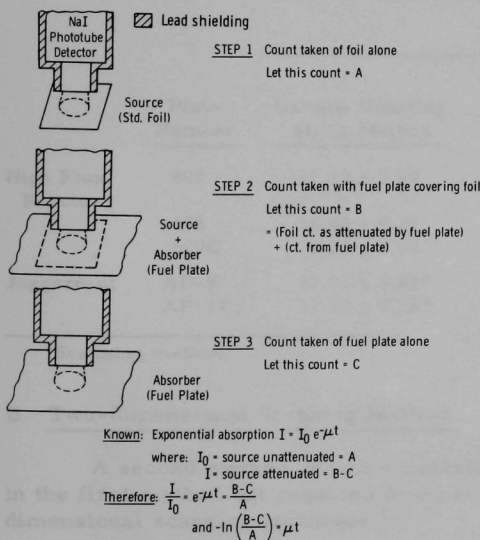


Figure 4

Experimental Procedure U²³⁵ Analysis Scintillation Spectrometry

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$$\frac{C_t}{C_0} = \frac{1 - e^{-\mu t}}{\mu t}$$

Figure 5

Self-absorption Relation for Exponentially Ab- sorbed Radiation

where:

C_0 = Activity of source with no self absorption

C_t = Activity of source with self absorption having thickness t and absorption coefficient μ

106-5990

The ratio of the corrected count rates for the standard foil and sample are equal to the ratio of their respective disintegration rates in the area under the collimator. The quantity of U^{235} in a unit area of the sample is then determined by direct proportion. The U^{235} content of the entire sample can be calculated by measuring the area of the sample and assuming the sample to be homogeneous. The area of clad samples may be measured from a radiograph. An example of the accuracy obtainable by this technique is given in Table I. The plates numbered 605, 598, and 500C were assayed by averaging the results of 15 counts taken over different areas on each plate.

Table I

COMPARISON OF DESTRUCTIVE AND NONDESTRUCTIVE ANALYSIS

		U^{235} Content, g		
	Plate Number	Gamma Counting, Static Method	Chemical and Mass-spectrometric Analysis	From Fabrication Data
High Flux Reactor	605	15.10 ± 0.30	15.07 ± 0.08	Not available
	598	18.43 ± 0.37	18.17 ± 0.11	Not available
	500C	4.24 ± 0.08	4.10 ± 0.02	Not available
Juggernaut	AD-9	$17.01 \pm 0.08^*$	16.953	17.01
	AP-11	$17.74 \pm 0.12^*$	17.735	16.68

*Scanning method

B. Two-dimensional Scanning Method

A second method, which eliminates two possible sources of error in the first method but requires more equipment, is based on a two-dimensional scanning technique.⁽²⁾

The technique is shown diagrammatically in Figure 6. A lathe, with the detector mounted on the cross feed, and the single-channel scintillation spectrometer used for scanning fuel plates are shown in Figure 7. The counting time is determined by the scanning speed of two orthogonal components of motion. In this method homogeneity need not be assumed and measurement of the area is not required. The resulting count accumulated during scanning is proportional to the area as well as the intensity of the source. The self-absorption corrections are calculated in the same manner as in the first method.

An example of a calibration curve obtained by two dimensional scanning for stainless steel-clad BORAX-V superheater plates with four

different core loadings is shown in Figure 8. The lower curve is the observed count rate as a function of core loading; the upper curve shows the count rate after correction for self-absorption as a function of core loading. The accuracy of the self-absorption correction is indicated by the fact that the points fall on a straight line passing through the origin.

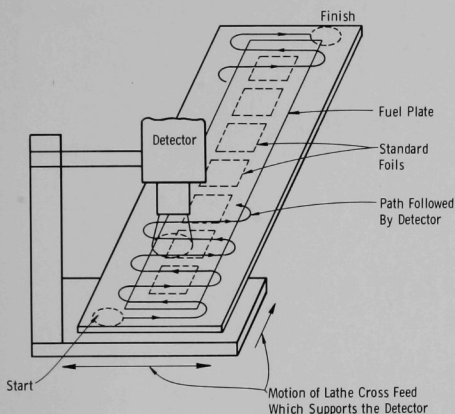
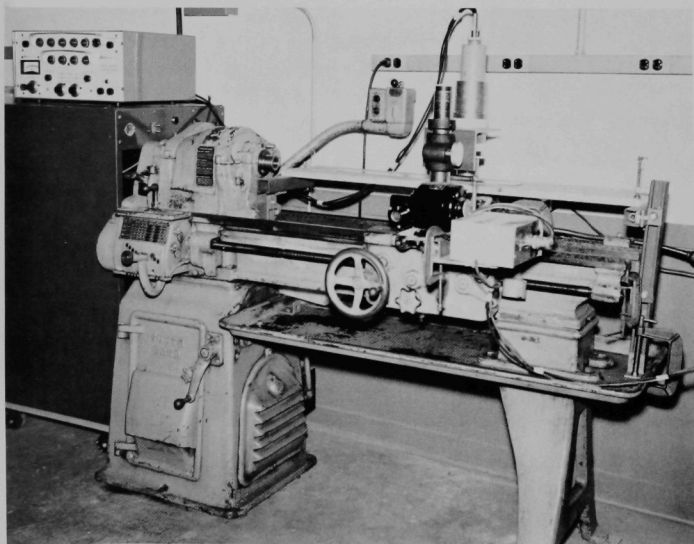


Figure 6

Total Scanning Technique U^{235}
Assay of Fuel Elements by
Scintillation Spectrometry

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Figure 7. Equipment for Two-dimensional Scanning of Fuel Plates

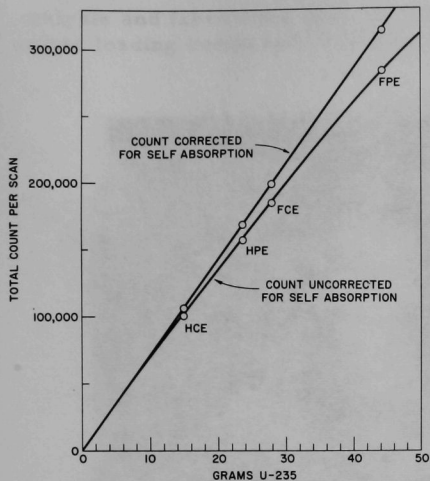


Figure 8
Calibration Curve Obtained
by Two-dimensional Scanning

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A modification of the two-dimensional scanning method has been applied to CP-5 type aluminum clad aluminum-uranium alloy tubes of 5.08- to 7.62-cm diameter. The tube is revolved on centers in a lathe while the detector scans along its length. A cylindrical lead absorber inside the tube permits the detector to measure gamma radiation from the detector side of the tube only. Standard foils mounted on a concentric cardboard tube are used to measure the self-absorption. A constant error of about 1% was encountered in this case. Further development would be necessary to make this procedure reliable for the tubular geometry.

III. DISCUSSION OF APPLICATIONS

If an entire reactor loading is to be assayed for U^{235} content, only a few plates need be standardized by one of the above methods. The balance of the plates can be compared with these standards. If the samples are small, they can be counted with a fixed geometry and the detector integrates the gamma flux over the area of the source. This technique gave satisfactory results for ALPR core blanks having dimensions of 17.463×8.41 cm when a 6.35-cm-diameter detector was used.⁽³⁾

The gamma flux from the entire area of a fuel plate can be integrated by the detector by moving the plate beneath the detector at a linear rate. The arrangement used for the Juggernaut loading and for BORAX-V superheater plates is shown in Figure 9. The larger detector in the figure was an alpha scintillator used for detecting radioactive surface contamination. Table I gives results obtained by scintillation spectrometry for individual fuel plates compared with chemical and mass spectrometer

analysis and fabrication data. Table II summarizes the results for an entire loading compared with fabrication data.⁽⁴⁾



106-5730

Figure 9. Equipment for Linear Scanning of Fuel Plates

Table II

MEASURED U^{235} LOADING COMPARED WITH FABRICATION DATA
FOR BORAX-V SUPERHEATER PLATES

Type of Loading	Number of Plates	U^{235} Content, g		
		Gamma Counting	Fabrication Data	% Deviation
HCE	186	2,797.66	2,783.57	+0.51
FCE	186	5,185.99	5,240.24	-1.04
HPE	226	5,334.21	5,341.89	-0.14
FPE	226	10,012.73	9,974.47	+0.38
Total	824	23,330.59	23,340.17	0.04

Fuel elements with large density-times-thickness products or complicated configurations cannot be standardized with uranium foils. If a large number of samples are to be nondestructively analyzed, it is usually

not objectionable to sacrifice several samples for destructive analysis. An alternative method of standardization is fabrication of standard fuel elements or simulated fuel elements. For example, standards for 0.0762-cm-diameter aluminum-uranium alloy wire were made by absorbing uranyl nitrate solution into nylon string of the same diameter. If the range of U^{235} content is not more than 5 to 10% from the average, two standards are usually sufficient for calibration. If the range is much larger a linear function cannot be assumed and additional standards are necessary.

IV. URANIUM DISTRIBUTION IN DISPERSION AND ALLOY FUELS

Distribution of uranium in dispersion or alloy-type fuels can be measured by scintillation spectrometry by counting small areas of the fuel and comparing the count rate with the average count rate. The average count rate for the sample can be determined by scanning the entire sample. For measuring the distribution of uranium it is advantageous to use the entire gamma spectrum, so that the desired accuracy can be obtained in the shortest counting time. A disadvantage of the technique is that thickness changes cannot be distinguished from inhomogeneity in the dispersion or alloy.

REFERENCES

1. White, F. K., and R. B. Perry, A Method of Determining Total U^{235} in Flat Configurations, COO-272 (Nov 1960).
2. Beyer, N. S., Nondestructive Fuel Assay with Emphasis on Scintillation Spectrometry, Paper No. 23, Nuclear Congress, 1962, Engineers Joint Council, 35 E. 47 St., New York 17, N. Y.
3. McGonnagle, W. J., and R. B. Perry, Analysis of Fuel Element Core Blanks for Argonne Low Power Reactor by Gamma Counting, ANL-5944 (Dec 1959).
4. Beyer, N. S., R. B. Perry, and B. Geren, Nondestructive Analysis by Radiation Detection of Fuel Content, Fuel Homogeneity, and Surface Contamination of the Superheater Plates for the BORAX-V Reactor, ANL-6746 (to be published).

not objectionable to sacrifice several samples for destructive analysis. An alternative method of standardization is fabrication of standard fuel elements or simulated fuel elements. For example, standards for 0.075-cm diameter aluminum-uranium alloy wire were made by absorbing uranyl nitrate solution into nylon string of the same diameter. If the range of U content is not more than 5 to 10% from the average, two standards are usually sufficient for calibration. If the range is much larger, a linear interpolation cannot be assumed and additional standards are necessary.

IV. URANIUM DISTRIBUTION IN DISPERSION AND ALLOY FUELS

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REFERENCES

1. White, E. K., and R. B. Perry, A Method of Determining Total U²³⁵ in Fuel Configurations, COO-175 (Nov 1959).
2. Beyer, N. S., Nondestructive Fuel Assay with Emphasis on Scintillation Spectrometry, Paper No. 53, Nuclear Congress, 1955, Engineers Joint Council, 35 E. 47 St., New York 17, N. Y.
3. McGonaggle, W. J., and R. B. Perry, Analysis of Fuel Element Core Blankets for Atomic Low Power Reactor by Gamma Counting, ANL-3944 (Dec 1957).
4. Beyer, N. S., R. B. Perry, and B. Geren, Nondestructive Analysis by Radiation Detection of Fuel Content, Fuel Homogeneity, and Surface Contamination of the Supracore Plates for the BORAX-V Reactor, ANL-5746 (to be published).

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